

NASA TT F-11,914

CALCULATION OF THE THERMODYNAMIC PROPERTIES OF
ALLOYS OF NICKEL AND COBALT

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Translation of "Raschet Termodinamicheskikh Svoystv
Splavov Nikelya i Kobal'ta"
Izvestiya Vysshikh Uchebnykh Zavedeniya,
Tsvetnaya Metallurgiya, Vol. 10, No. 4, pp. 35-39, 1967

FACILITY FORM 602	N 68-35224	
	(ACCESSION NUMBER)	(THRU)
	<u>10</u>	<u>1</u>
	(PAGES)	(CODE)
	<u>✓</u>	<u>17</u>
	(NASA CR OR TMX OR AD NUMBER)	(CATEGORY)



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ABSTRACT. The theory of regular solutions is applied to the problem of determining component activity, thermodynamic quantities and heats of mixing of strongly interacting systems. Formulas are produced for calculation of thermodynamic and thermochemical properties of solutions consisting of components which form large negative and positive asymmetrical deflections from the law of ideal solutions. The intermolecular interaction constants are calculated for many alloys of nickel and cobalt with various elements, which can be used to estimate the concentration dependence of the activity of the components, the mixing heat and the area of existence of two immiscible phases.

Thermodynamic quantities are not only of theoretical interest, since they /35* allow us to evaluate the nature of the interatomic interaction, but also are of practical value in the determination of gas saturation [1], diffusion processes [2], and furthermore are necessary for the solution of certain technological problems in the production of nonferrous metals [3].

In order to determine the activity of components, the partial and integral thermodynamic quantities and the heats of mixing of strongly interacting systems, the equilibrium between the melt and a chemical compound of practically constant composition near the maximum of the liquidus curve is used; the principles of this method in the framework of the theory of ideal solutions are analyzed in monograph [4]. In the present work, we shall extend the method by applying the theory of regular solutions as well as expressions which take into consideration the asymmetry of the thermodynamic properties.

On the basis of [4], we have the following expression for the points of the maximum

$$\left(\frac{d^2 T}{dx_2^2} \right)_{\max} = - \frac{T}{\Delta H (1 - x_2)} \left(\frac{d \mu_2}{dx_2} \right)_{\max}, \quad (1)$$

where μ_2 and x_2 are the chemical potential and atomic fraction of the component, T and ΔH are the melting temperature and heat of the chemical

*Numbers in the margin indicate pagination in the foreign text.

compound.

From the theory of regular solutions we have

$$\mu_2 = \mu_2^0 + RT \ln x_2 + (1-x_2)^2 Q,$$

then after differentiation we find

$$\left(\frac{d^2 T}{dx_2^2} \right)_{\max} = - \frac{RT^2}{\Delta H x_2 (1-x_2)} + \frac{2QT}{\Delta H}$$

or

$$Q = \left(\frac{d^2 T}{dx_2^2} \right)_{\max} \frac{\Delta H}{2T} + \frac{RT}{2x_2(1-x_2)}, \quad (2)$$

where Q is the energy of the interchange in the system consisting of components 1 and 2.

Considering an equation from [5]

$$\mu_2 = \mu_2^0 + RT \ln x_2 + (Q_1 + 2Q_2 + 3Q_3) x_1^2 - (2Q_2 + 6Q_3) x_1^3 + 3Q_3 x_1^4 \quad (3) \quad \underline{/36}$$

after differentiation and substitution in expression (1), we produce

$$\begin{aligned} \left(\frac{d^2 T}{dx_2^2} \right)_{\max} = & - \frac{RT^2}{\Delta H x_2 (1-x_2)} + \frac{2T(Q_1 + 2Q_2 + 3Q_3)}{\Delta H} - \\ & - \frac{6T(Q_2 + 3Q_3)(1-x_2)}{\Delta H} + \frac{12TQ_3(1-x_2)^2}{\Delta H}. \end{aligned} \quad (4)$$

The value of ΔH is determined from experimental data or calculated from the additive nature of the melting entropy of the pure components [6]; the numerical values of the heats and temperatures of melting of the components are taken from handbook data [7, 8].

The change in temperature near the maximum is described by a parabola like

$$T = T_0 + a(x_2 - x_2^m)^2 + b(x_2 - x_2^m)^3, \quad (5)$$

where x_2^m is the concentration of component 2, corresponding to the maximum on the liquidus curve;

a and b are constants defined from experimental data [9].

The results of calculations using equations (2) and (5) are presented together with the initial data in Table 1. The values of the heats and temperatures of melting of nickel and cobalt are taken as 17.7 and 15.7 kJ/g·atom, 1455 and 1495° respectively [7].

In nickel melts with Be, Ce, La, Sb, Th, Zr and Co with Si, an asymmetrical deflection is noted, indicating systematic changes in Q for the various compositions. This confirms once again the data of work [5], in which the complex nature of the change in activity with melt composition is noted.

In this case, we must use equations like (3) and (4). Combined solution of three equations (4) for the three compounds allows us to determine the constants Q_1 , Q_2 and Q_3 . For the system Ni-Ce, this determination gives us $Q_1 = -187$; $Q_2 = 254$ and $Q_3 = -156.5$ kJ/g·atom. The activity coefficients for 1600° are determined [5] by the expressions

$$\begin{aligned} \lg f_{\text{Ni}} &= -12,3 x_{\text{Ce}}^2 + 22,9 x_{\text{Ce}}^3 - 13,1 x_{\text{Ce}}^4, \\ \lg f_{\text{Ce}} &= -4,15 x_{\text{Ni}}^2 + 12,0 x_{\text{Ni}}^3 - 13,1 x_{\text{Ni}}^4. \end{aligned}$$

The results of calculations considering $a = fx$ (Figure 1) indicate a considerable interaction in this system, and if the components are mixed, a considerable quantity of heat is liberated.

In recent years, rare metals are being ever more frequently used for alloying, desulfuration and gas removal, but there is no information at all concerning the mixing heats. However, these data are required for the composition of thermal balances and are the basis for automation of the thermal operation of an aggregate. Calculations of the thermochemical properties can be performed on the assumption that the excess entropy is zero, i.e., by assuming

Table

Calculation of value of Q using equation (2)

System	ΔH_o^B , kJ/g·at	Compound			$t, ^\circ\text{C}$	ΔH^m , kJ/g·at	Equation of liquidus line, $t, ^\circ\text{C}$	Q, kJ/g·at
		Formula	x_2^m	x_2^m				
Ni-Al	10,3	NiAl	0,50	0,50	1638	20,4	$t = 1638 - 6800(x_{Al} - x_{Al}^m)^2 - 11500(x_{Al} - x_{Al}^m)^3$	-41,5
Ni-Be	9,6	NiBe	0,50	0,50	1472	14,3	$t = 1472 - 10200(x_{Be} - x_{Be}^m)^2 - 65000(x_{Be} - x_{Be}^m)^3$	-54,7
"	9,6	Ni ₅ Be ₂₁	0,808	0,808	1264	10,7	$t = 1264 - 3550(x_{Be} - x_{Be}^m)^2 + 77000(x_{Be} - x_{Be}^m)^3$	+16,4
Ni-Ce	8,9	Ni ₅ Ce	0,167	0,167	1315	15,8	$t = 1315 - 21500(x_{Ce} - x_{Ce}^m)^2 - 49000(x_{Ce} - x_{Ce}^m)^3$	-166,5
"	8,9	NiCe	0,50	0,50	670	8,7	$t = 670 - 4800(x_{Ce} - x_{Ce}^m)^2 - 32500(x_{Ce} - x_{Ce}^m)^3$	-28,6
"	8,9	NiCe ₃	0,75	0,75	485	6,6	$t = 485 - 3300(x_{Ce} - x_{Ce}^m)^2 + 2970(x_{Ce} - x_{Ce}^m)^3$	-11,9
Ni-Ge	33,8	NiGe _{0,54}	0,35	0,35	1200	24,2	$t = 1200 - 17500(x_{Ge} - x_{Ge}^m)^2 - 29500(x_{Ge} - x_{Ge}^m)^3$	-261
Ni-La	11,7	Ni ₅ La	0,167	0,167	1325	16,3	$t = 1325 - 12000(x_{La} - x_{La}^m)^2 + 46200(x_{La} - x_{La}^m)^3$	-74,8
"	11,7	NiLa	0,50	0,50	685	9,6	$t = 685 - 16500(x_{La} - x_{La}^m)^2 - 111000(x_{La} - x_{La}^m)^3$	-149,5
"	11,7	NiLa ₃	0,75	0,75	515	7,8	$t = 515 - 5300(x_{La} - x_{La}^m)^2 + 42200(x_{La} - x_{La}^m)^3$	-41,0
Ni-Mg	9,0	Ni ₂ Mg	0,333	0,333	1145	14,3	$t = 1145 - 3600(x_{Mg} - x_{Mg}^m)^2 + 11500(x_{Mg} - x_{Mg}^m)^3$	-9,7
Ni-Nb	25,2	Ni ₃ Nb	0,25	0,25	1403	16,7	$t = 1403 - 19000(x_{Nb} - x_{Nb}^m)^2$	-153,2
Ni-Pr	10,0	Ni ₅ Pr	0,167	0,167	1365	16,3	$t = 1365 - 13000(x_{Pr} - x_{Pr}^m)^2 + 46700(x_{Pr} - x_{Pr}^m)^3$	-80,4
"	10,0	NiPr	0,50	0,50	730	9,3	$t = 730 - 21000(x_{Pr} - x_{Pr}^m)^2 + 217000(x_{Pr} - x_{Pr}^m)^3$	-178,0
"	10,0	NiPr ₃	0,75	0,75	533	7,1	$t = 533 - 16500(x_{Pr} - x_{Pr}^m)^2 + 109200(x_{Pr} - x_{Pr}^m)^3$	-127,4
Ni-Pu	8,2	Ni ₅ Pu	0,167	0,167	1435	17,2	$t = 1435 - 37000(x_{Pu} - x_{Pu}^m)^2$	-320,3
Ni-Sb	20,1	Ni ₅ Sb ₂	0,286	0,286	1162	19,8	$t = 1162 - 12300(x_{Sb} - x_{Sb}^m)^2 + 8400(x_{Sb} - x_{Sb}^m)^3$	-140,6
"	20,1	NiSb	0,50	0,50	1153	23,3	$t = 1153 - 4500(x_{Sb} - x_{Sb}^m)^2$	-49,8
Ni-Si	50,6	Ni ₂ Si	0,333	0,333	1318	26,8	$t = 1318 - 15000(x_{Si} - x_{Si}^m)^2 - 53500(x_{Si} - x_{Si}^m)^3$	-222,9
"	50,6	NiSi	0,50	0,50	992	25,5	$t = 992 - 13500(x_{Si} - x_{Si}^m)^2 + 105000(x_{Si} - x_{Si}^m)^3$	-251
Ni-Sn	7,2	Ni ₃ Sn	0,25	0,25	1174	16,3	$t = 1174 - 6200(x_{Sn} - x_{Sn}^m)^2 - 130000(x_{Sn} - x_{Sn}^m)^3$	-37,8
"	7,2	Ni ₃ Sn ₂	0,40	0,40	1264	18,2	$t = 1264 - 6400(x_{Sn} - x_{Sn}^m)^2$	-49,2
Ni-Ta	24,7	Ni ₃ Ta	0,25	0,25	1545	17,4	$t = 1545 - 15300(x_{Ta} - x_{Ta}^m)^2 - 53600(x_{Ta} - x_{Ta}^m)^3$	-106

Table
(continued)

Calculation of value of Q using equation (2)

System	ΔH_f^B , kJ/g·at	Compound			ΔH_f^m , kJ/g·at	Equation of liquidus line, t. °C	Q, kJ/g·at
		Formula	x_2^m	t, °C			
Ni-Th	15,7	Ni ₅ Th	0,167	1530	17,8	$t = 1530 - 26\,000(x_{Th} - x_{Th}^m)^2 + 85\,000(x_{Th} - x_{Th}^m)^3$	-202,6
"	15,7	NiTh	0,50	1200	13,4	$t = 1200 - 8000(x_{Th} - x_{Th}^m)^2 + 20\,000(x_{Th} - x_{Th}^m)^3$	-34,6
Ni-Ti	18,8	NiTi	0,50	1240	14,9	$t = 1240 - 10\,300(x_{Ti} - x_{Ti}^m)^2 + 27\,500(x_{Ti} - x_{Ti}^m)^3$	-76,1
"	18,8	NiTi	0,50	1310	15,5	$t = 1310 - 14\,000(x_{Ti} - x_{Ti}^m)^2 + 45\,500(x_{Ti} - x_{Ti}^m)^3$	-110,8
"	18,8	Ni ₃ Ti	0,25	1378	16,6	$t = 1378 - 18\,800(x_{Ti} - x_{Ti}^m)^2 + 11\,300(x_{Ti} - x_{Ti}^m)^3$	-152,4
"	18,8	Ni ₃ Ti	0,25	1380	16,6	$t = 1380 - 12\,000(x_{Ti} - x_{Ti}^m)^2$	-84,1
Ni-Zr	19,2	NiZr ₂	0,667	1200	13,9	$t = 1200 - 28\,000(x_{Zr} - x_{Zr}^m)^2$	-236,4
"	19,2	Ni ₃ Zr	0,25	1740	20,0	$t = 1740 - 11\,000(x_{Zr} - x_{Zr}^m)^2$	-64,7
Co-Al	10,3	CoAl	0,50	≈ 1645	19,2	$t = 1645 - 5500(x_{Al} - x_{Al}^m)^2 - 19\,000(x_{Al} - x_{Al}^m)^3$	-23,1
Co-Be	9,6	CoBe	0,50	1505	13,4	$t = 1505 - 10\,000(x_{Be} - x_{Be}^m)^2 - 32\,000(x_{Be} - x_{Be}^m)^3$	-45,8
Co-Ce	8,9	CoCe ₃	0,75	≈ 480	6,3	$t = 480 - 7200(x_{Ce} - x_{Ce}^m)^2 - 8600(x_{Ce} - x_{Ce}^m)^3$	-43,5
Co-Ge	33,8	Co ₂ Ge	0,333	1200	22,4	$t = 1200 - 10\,000(x_{Ge} - x_{Ge}^m)^2 + 38\,500(x_{Ge} - x_{Ge}^m)^3$	-124,5
Co-Sb	20,1	CoSb _{0,785}	0,44	≈ 1195	21,8	$t = 1195 - 2900(x_{Sb} - x_{Sb}^m)^2 + 2500(x_{Sb} - x_{Sb}^m)^3$	-17,8
Co-Si	50,6	Co ₂ Si	0,333	1332	25,6	$t = 1332 - 15\,000(x_{Si} - x_{Si}^m)^2 + 73\,000(x_{Si} - x_{Si}^m)^3$	-209,1
"	50,6	CoSi	0,50	1460	33,7	$t = 1460 - 11\,000(x_{Si} - x_{Si}^m)^2 + 42\,000(x_{Si} - x_{Si}^m)^3$	-185,2
"	50,6	CoSi ₂	0,667	1326	36,8	$t = 1326 - 5300(x_{Si} - x_{Si}^m)^2 + 5300(x_{Si} - x_{Si}^m)^3$	-91,9
Co-Sn	7,2	CoSn _{0,51}	0,35	1170	15,3	$t = 1170 - 2100(x_{Sn} - x_{Sn}^m)^2 + 5200(x_{Sn} - x_{Sn}^m)^3$	+4,1
Co-U	12,6	Co ₂ U	0,333	1170	12,9	$t = 1170 - 5000(x_{U} - x_{U}^m)^2 - 19\,000(x_{U} - x_{U}^m)^3$	-17,7

$$\Delta H^{\text{mix}} = \Delta G^{\text{ex}}, \quad (6) \quad /37$$

where ΔG^{ex} is the excess molar Gibbs energy, determined on the basis of the data of [5] by expression

$$\Delta G^{\text{mix}} = Q_1 x_1 x_2 + Q_2 x_1 x_2^2 + Q_3 x_1 x_2^3. \quad (7)$$

The greatest errors introduced by assumption (6) should be observed for melts with high values of Q . For example, in melts of nickel with silicon, the maximum values of the functions are $\Delta G^{\text{ex}} = -46 \text{ kJ/g}\cdot\text{atom}$ [10], and $\Delta H^{\text{mix}} = -58 \text{ kJ/g}\cdot\text{atom}$ [10, 11], from which, on the basis of

$$\Delta G^{\text{ex}} = \Delta H^{\text{mix}} - T \Delta S^{\text{ex}}$$

at 1600° , the value of $\Delta S^{\text{ex}} = -6.4 \text{ j/g}\cdot\text{atom}\cdot\text{deg}$. As we can see, the error

introduced by the assumption that $\Delta S^{\text{ex}} = 0$ is 20%, which is greater than the accuracy of the calculations. For weakly interacting systems, the results of calculations from the state diagram (Co-Cu and Ni-Cu melts [5]) are confirmed by direct calorimetric measurements of the mixing heats of nickel and cobalt with copper [12, 13].

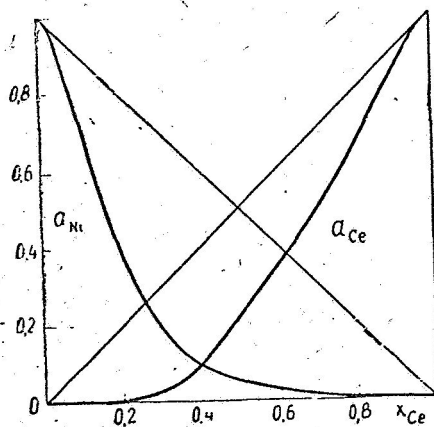


Figure 1. Activity A_s as a Function of Concentration in Ni-Ce Alloys at 1600°

For the system Co-Si, we produce from equation (4) $Q_1 = -349.5$; $Q_2 = +213.4$ and $Q_3 = -24.2 \text{ kJ/g}\cdot\text{atom}$, from which, on the basis of (6) and (7), we can produce values of ΔH^{mix} and ΔG^{ex} . Calculations show that the maximum of the functions is located at $x_{\text{Si}} = 0.408$, which corresponds to the experimental data of [14, 15], but the numerical values of the calculated quantities are somewhat higher than the experimental values.

In order to test other values of Q , we can note that the value of $Q_{\text{Ni-Ti}}$ presented in work [16] almost corresponds with the minimal values for NiTi Ni_3Ti from Table 1. In Ni-Al and Co-Al

alloys, a negative deflection is noted, and the results of the calculations qualitatively correspond with the experimental data [17].

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TABLE 2. CALCULATION OF VALUES OF Q_1 AND Q_2 FROM EQUATIONS (8) AND (9)

System	$t, ^\circ\text{C}$	Phase Composition		$Q_1, \text{kJ/g}\cdot\text{at.}$	$Q_2, \text{kJ/g}\cdot\text{at.}$
		x_2'	x_2''		
Ag—Ni	1435	0,02	0,97	57,7	—5,3
Ag—Ni*	1435	0,0389	0,978	56,7	—7,5
Co—Bi	1345	0,02	0,823	53,4	—30,0
Co—Se	1448	0,034	0,31	—1,0	—138,4
Co—Pb*	1438	$\approx 0,0067$	0,9933	72,0	—
Ni—Ti	1387	0,025	0,96	53,4	—5,9
Ni—Pb	1340	0,127	0,72	35,2	—10,0
Ni—Pb*	1340	0,127	0,79	35,2	—6,1

For systems with positive deflection, the constants Q , which allow us to determine the thermodynamic and thermochemical properties of the alloys, are determined from the area of immiscibility [9]. The compositions of the phases are determined by the equality of the chemical potentials of the components in both phases (phase numbers marked with primes)

$$\mu_1' = \mu_1'' \text{ and } \mu_2' = \mu_2'',$$

from which after substitution of expressions (3) for $Q_3 = 0$ we produce the two equations

$$RT \ln \frac{x_1'}{x_2'} + [(x_2')^2 - (x_2'')^2] (Q_1 - Q_2) + [(x_2')^3 - (x_2'')^3] 2Q_2 = 0, \quad (8)$$

$$RT \ln \frac{x_2''}{x_1''} + [(x_1'')^2 - (x_1')^2] (Q_1 + 2Q_2) - [(x_1'')^3 - (x_1')^3] 2Q_2 = 0, \quad (9)$$

joint solution of which allows us to determine Q_1 and Q_2 . Calculations of alloys of cobalt and nickel with various elements [9] are presented in Table 2, where the asterisks mark systems with boundaries of termination of miscibility clarified by the data of [18-21].

Solving equations (8) and (9) for the temperature and fixing of the composition of one phase, we can determine by selection the composition of the second phase for which the values of temperatures from these equations correspond. The critical composition and temperature are determined from the expressions

$$x_2^{cr} = -\frac{Q_1 - 4Q_2}{9Q_2} - \sqrt{\left(\frac{Q_1 - 4Q_2}{9Q_2}\right)^2 + \frac{Q_1 - Q_2}{9Q_2}} \quad (10)$$

and

$$T_{cr} = \frac{2(Q_1 - Q_2)}{R} x_2^{cr} (1 - x_2^{cr}) + \frac{6Q_2}{R} (x_2^{cr})^2 (1 - x_2^{cr}). \quad (11)$$

An example of the calculation of the area of immiscibility is shown on Figure 2 for the system Ni-Pb.

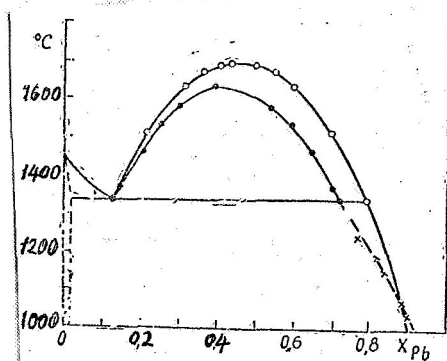


Figure 2. Diagram of State of System Ni-Pb.

x -- Experimental data of [20]; o -- Calculations using equations (8)-(11)

Using the values of Q which we have found, we can calculate the properties of multi-component systems such as are the real alloys, using the equations produced in works [16, 22]. /39

Conclusions

1. Formulas are produced for calculation of thermodynamic and thermochemical properties of solutions consisting of components which form large negative and positive asymmetrical deflections from the law of ideal solutions.

2. The intermolecular interaction constants are calculated for many alloys of nickel and cobalt with various

elements, which can be used to estimate the concentration dependence of the activity of the components, the mixing heat and the area of existence of two immiscible phases.

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